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World-Wide Distribution of Natural Radiocarbon*

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The natural radiocarbon content of biological materials collected from locations widely scattered over the earth's surface have shown no evidence of the latitudinal variation which would be expected were the turnover time for the biosphere comparable with the mean life of radiocarbon. The strong dependence of the intensity of the cosmic-ray neutrons, which produce the natural radiocarbon, on latitude would be expected to cause this effect. Since it is expected, however, on geochemical evidence that the turnover time for the biosphere would be very short compared to the mean life of radiocarbon, the observed uniformity is not surprising.

The observed mean assay of carbon in equilibrium with the biosphere is 16.1±0.5 dpm/g C. Consideration of the amounts of the various types of carbon in equilibrium with the biosphere, of which the most abundant is the inorganic carbonate and bicarbonate ions dissolved in sea water, gives 8.3 grams per cm^2 of the earth's surface. A direct comparison with observed cosmic-ray neutron intensity is made by taking the observed thermal intensity and correcting according to standard formulas for the epithermal production of radiocarbon, to give a total of 156±30 neutrons/cm^2 of earth's surface/minute on the average. Therefore, one expects that the mean specific activity of carbon in equilibrium with the biosphere would be the ratio of 156 to 8.3, or 18.8±5 dpm/g. The agreement between the predicted and observed specific activity is gratifying, but owing to the considerable uncertainties in our knowledge of the cosmic-ray intensity and the amount of diluting carbon, it may be that it is somewhat fortuitous. It does appear to indicate, however, that no considerable variation in cosmic-ray intensity over the past several thousand years has occurred. The fact that the principal part of the diluting reservoir is inorganic material in the sea water serves to make any large variation in the amount of carbon in this reservoir unlikely.

I. INTRODUCTION

THE occurrence of radiocarbon in nature because of cosmic-ray action on the atmosphere[1-5] and its use for archaeological and geological dating[6,7] raise several points of general interest. One of these is the world-wide distribution of radiocarbon and the balance between the cosmic radiation intensity and the observed specific radioactivity of living matter.[1,4,6]

The cosmic-ray primaries produce neutron secondaries in the intensity $\bar{Q}$ (neutrons/cm^2/sec) as averaged over the earth's surface. Since no other process for neutron absorption in air is likely to utilize more than 2 or 3 percent as many neutrons as the $N^{14}(n,p)C^{14}$ reaction to produce radiocarbon, we immediately conclude that the terrestrial inventory of radiocarbon must be such as to insure a disintegration rate of $5.1\times10^{18}\bar{Q}$, where 5.1×10^{18} is the area of the earth in cm^2. This assumes that the process has been occurring at a steady value of $\bar{Q}$ for several half-lives, or sometime over 20,000 years, so that a steady-state condition has been established in which the rates of formation and disintegration are equal.

The steady-state inventory must be $(5.1\times10^{18}\bar{Q}\times14)/(6\times10^{23}\lambda)$ grams, where λ is the disintegration rate corresponding to the half-life of 5568±30 years.[8-12] The measured value of $\bar{Q}$ appears to be close to 2.6 neutrons/cm^2/sec, as we shall see later. These figures correspond to an inventory of 80 metric tons of radiocarbon and an annual production rate of 9.8 kg.

The questions of the world-wide distribution of radiocarbon and the expected average specific radioactivity for the various forms of terrestial carbon involve two principal considerations. First, the neutrons are absorbed at high altitudes,[13] so the source of radiocarbon can be considered to be at 30,000 ft or higher and the newly born carbon atoms will probably burn to CO_2 before reaching the earth. Therefore, we are led to expect all plant life to be radioactive, since the atmospheric CO_2 on which it lives is radioactive and the animals living off plants must possess the same radioactivity. In addition we see that inorganic carbon such as CO_3^{--}, HCO_3^-, and H_2CO_3 in the sea which exchange with CO_2 will be radioactive. This last category proves to constitute the main part of the diluting carbon inventory.

The second major consideration is whether the mixing is likely to be adequate in the lifetime of the average radiocarbon atom (8033 years, average life). The mixing times for the atmosphere and the biosphere seem certain to be short as compared with 8033 years,

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[1] W. F. Libby, Phys. Rev. **69**, 671 (1946).

[2] Anderson, Libby, Weinhouse, Reid, Kirshenbaum, and Grosse, Science **105**, 576 (1947).

[3] A. V. Grosse and W. F. Libby, Science **106**, 88 (1947).

[4] Anderson, Libby, Weinhouse, Reid, Kirshenbaum, and Grosse, Phys. Rev. **72**, 931 (1947).

[5] E. C. Anderson, Doctoral Thesis, University of Chicago, 1949.

[6] Libby, Anderson, and Arnold, Science **109**, 227 (1949).

[7] J. R. Arnold and W. F. Libby, Science **110**, 678 (1949).

[8] Engelkemeir, Hamill, Inghram, and Libby, Phys. Rev. **75**, 1825 (1949).

[9] W. M. Jones, Phys. Rev. **76**, 885 (1949).

[10] Miller, Ballentine, Bernstein, Friedman, Nier, and Evans, Phys. Rev. **77**, 714 (1950).

[11] Engelkemeir and Libby, Rev. Sci. Insrr. **21**, 550 (1950).

Note added in proof: The 5568 year half-life is derived from a correction[11] on our earlier value of 5720[8] and an averaging with other published values.

[12] Hawkings, Hunter, Mann, and Stevens, Can. J. Research, **27**, 545 (1949).

[13] For example, L. C. L. Yuan, Phys. Rev. **77**, 728 (1950).

but the status of ocean mixing is less certain. It seems likely, however, that this probably is essentially complete also.

The experimental values for the inventory are given in Table I.

The experimental value for the specific radioactivity of living (biospheric and organic) material is 15.3±0.5 disintegrations per minute per gram of carbon; for inorganic carbonate carbon, 16.2±0.5; to give 16.1±0.5 for the average carbon in the inventory, which weighs 8.3 g/cm^2. Multiplying, we have 134 dpm/cm^2 *vs.* 156±30 cosmic-ray neutrons/cm^2-min., the rate expected for a complete steady-state balance. Since the data are hardly more accurate than twenty percent, the check seems satisfactory. In addition, no significant variation with latitude, longitude, altitude, or depth in the sea has been found, so the mixing seems to be essentially complete.

II. NEUTRON PRODUCTION RATE, Q

The distribution of neutrons[13–18] in the atmosphere is exponential up to about 40,000 feet with a half-thickness of about one meter of water equivalent. Above 40,000 feet a maximum is reached followed by a rapid decrease indicating the secondary nature of the neutrons. In addition to the altitude dependence, the neutron intensity shows a strong dependence on geomagnetic latitude,[14,17] the intensity at high latitudes being about four times that at the magnetic equator. This variation must be considered, of course, in calculating the world-wide average intensity, $\bar{Q}$.

The measurements on neutron intensity which we use for our calculations of total intensity have been on the thermal component as defined by cadmium absorption for a boron detector. We shall take this to mean that that fraction of the neutron spectrum lying below 0.4 ev has been measured, and in order to calculate Q and its world-wide average $\bar{Q}$, we must determine theoretically the probable ratio of the total intensity to the thermal intensity. Bethe, Korff, and Placzek[19] and Placzek[20] have considered the problem in detail. Following their treatment and using the later data of Melkonian,[21] together with the data collected in the M.I.T. Volume,[22] we shall assume that in the range 0.4 ev to 0.5 Mev the total cross section in units of 10^{-24} cm^2 for air is given by

$$\sigma_t = 8.56 + (0.266/E^{\frac{1}{2}}),$$

and that the capture cross section is given by

$$\sigma_c = 0.266/E^{\frac{1}{2}}.$$

The expression for the ratio of the total number of neutrons generated to the number which reach the thermal range is e^y, where y is given by

$$y = (2/0.248)\int_{0.4}^{5\times 10^5} \sigma_c dE/\sigma_t E,$$

where the number 0.248 is the fractional energy loss in elastic collision with the nitrogen atom, the bond strength being neglected. Integration and substitution give 0.79 for the integral and the calculated ratio of total production to thermal population of 2.20. It is clear, however, that one must consider the effect of the very strong bonds in the nitrogen and oxygen molecules on the cooling process. Study of the analogous problem of the effect of binding on the neutron elastic and inelastic cross sections for hydrogen[23] leads us to expect that no large error is made in neglecting the binding for cooling down to the Cd cut-off of 0.4 ev, which is 1.5 vibrational quanta for the average air molecule.

It is necessary to make a correction for the absorption above 0.5 Mev. It has been shown by Barschall and Battat[24] and others[22] that resonance production of radiocarbon occurs in this range at cross sections rising to maxima of 0.1×10^{-24} cm^2 and falling to about 0.01×10^{-24} cm^2 in the range 0.5 Mev to 2 Mev. To obtain a probable upper limit for the capture contribution in this high energy range, we assume that only elastic scattering occurs and an average capture cross section of 0.035×10^{-24} cm^2 applies throughout the high energy interval. This gives an additional contribution of 7 percent to the ratio of total to thermal neutron population. The corrected ratio is 2.36.

The value of Q is now to be obtained at any given latitude by multiplying the observed thermal intensity Q by the number 2.36. For this purpose we choose the data of Yuan and Ladenberg[13–15] obtained at Princeton, New Jersey, which give 1.9 as the number of slow cosmic-ray neutrons absorbed per sec per cm^2 of earth's surface at that latitude (we are very obliged to Professor Ladenberg for making this result available to us before publication). Using Simpson's data for the variation of Q with latitude and integrating over the surface of the earth, we find for the average thermal flux, 1.1 thermal cosmic-ray neutrons/cm^2-sec. Finally, multiplying by the ratio of total production to thermal neutron population, we obtain 2.6 as the most likely value for $\bar{Q}$, the

TABLE I. Carbon inventory.

Source	Amount
Ocean "carbonate"	7.25 g/cm^2
Ocean, dissolved organic	0.59
Biosphere	0.33
Atmosphere	0.12
Total	8.3 g/cm^2

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[14] L. C. L. Yuan, Phys. Rev. **76**, 1267, 1268 (1949).

[15] L. C. L. Yuan and R. Ladenburg, Bull. Am. Phys. Soc. **23**, No. 2, 21 (1948).

[16] L. C. L. Yuan, Phys. Rev. **74**, 504 (1948).

[17] J. A. Simpson, Jr., Phys. Rev. **73**, 1389 (1948).

[18] Agnew, Bright, and Froman, Phys. Rev. **72**, 203 (1947). (This paper contains references to many earlier measurements.)

[19] Bethe, Korff, and Placzek, Phys. Rev. **57**, 573 (1940).

[20] G. Placzek, Phys. Rev. **69**, 423 (1946).

[21] E. Melkonian, Phys. Rev. **76**, 1750 (1949).

[22] *Science and Engineering of Nuclear Power* (Addison-Wesley Press, Inc., 1947), Vol. 1, pp. 408–411.

[23] H. A. Bethe, Revs. Modern Phys. **9**, 122–127 (1937).

[24] H. H. Barschall and M. E. Battat, Phys. Rev. **70**, 245 (1946).

average total production of cosmic-ray neutrons per cm^2 of earth's surface per sec. Considering the various sources of error in this figure, it seems that it probably is good to about 20 percent, though there is, of course, considerable possibility that the error is somewhat larger or smaller than this.

III. EXPECTED TERRESTRIAL DISTRIBUTION AND SPECIFIC ACTIVITY OF LIVING MATTER

Turning now from the production of radiocarbon to the process of dilution, we consider the geochemical fate of the C^{14} produced by the cosmic-ray neutrons. It is clear that it will be rapidly converted to carbon dioxide and therefore will be diluted by all carbon dioxide in the air. It then follows, of course, that all plant life and all animal life on earth must contain radiocarbon, and that the carbonate dissolved in the sea, which will exchange with atmospheric carbon dioxide, will also be radioactive. We shall find that the largest part of the diluting material is the carbonate dissolved in the sea.

In order to estimate the amount of dissolved carbonate in the sea one needs to know the alkalinity and the $p\mathrm{H}[-\log(H^+)]$ of the ocean water. The alkalinity is the excess of positive ions over the anions of strong acids, and is made up by the ionization of weak acids, of which carbonic acid is the principal one in the ocean. The average value of the alkalinity is well established at 2.43 milliequivalents per liter.[25] This must be reduced to 2.37 for the boric acid in the water. The total amount of dissolved inorganic carbon in the sea is not uniquely determined by the alkalinity, owing to the possibility of its existence as H_2CO_3, HCO_3^-, and $CO_3^=$. The ratio of the amounts in these various forms is determined by the pH.

The values of the dissociation constants for H_2CO_3 and HCO_3^- have been carefully measured by a number of investigators[26–28] as functions of pH, salinity, temperature, and pressure in both natural and artificial sea water. The results appear to be very reliable. Using standard data we find for average ocean water $K_1=1.26\times10^{-6}$ and $K_2=7.4\times10^{-10}$. The variation in the pH of the ocean is surprisingly small, the average value[29] being 8.0 and the total variation less than 0.1 pH unit. The application of these numbers gives 26.2 mg of carbon per kg of sea water, or 7.25 g/cm^2 of the earth's surface, corresponding to a total mass of 3.7×10^{19} grams. The error in this figure may be as large as 10 percent, arising mainly from the small but significant observed variation of the pH and the scatter in the values for the alkalinity as reported by various observers.

The remaining items in the inventory of exchangeable carbon are the dissolved organic carbon in the sea, the atmospheric carbon dioxide, and the biosphere. For the first of these the best figure[30] appears to be 2 mg/liter, corresponding to 0.59 g/cm^2.

Atmospheric carbon dioxide has been determined by several people. The values are: Buch,[31] 0.12 (polar) to 0.13 (tropical and continental); Paneth,[32] 0.12; and Vernadsky,[33] 0.12. We will use 0.12 g/cm^{-2} for the amount of carbon in atmospheric carbon dioxide. This is equivalent to CO_2 partial pressure of 0.21 mm Hg, or a concentration of 0.028 percent.

The amount of carbon in living matter is difficult to estimate and appears to have been grossly overestimated by some writers in the past. We base our estimate on the rate of fixation of carbon by photosynthesis, a quantity which appears to be fairly well established.[34] Fortunately it can be shown that biosphere carbon is small compared with ocean carbonate, and therefore an error in its estimation will not be of great importance.

The total rate of fixation of carbon by land plants has been investigated by Schroeder[35] who concluded that the average annual fixation by this source is 1.63×10^{16} grams or 3.2 mg/cm^{-2}. Riley[36] made an investigation of the fixation by ocean plankton and arrived at a figure of 30.5 mg/cm^{-2} for the average annual rate. Rabinowitch[37] estimates that on the basis of the solar energy flux, reflection losses, and photosynthetic efficiency, not more than 60 mg/cm^{-2} could be fixed annually, indicating that Riley's figure cannot be too low by any large factor. We shall use the sum of the values given by Schroeder and Riley as the total annual rate of fixation of carbon, namely 33 mg/cm^{-2}. The total amount of carbon contained in the biosphere at any time will be given by the fixation rate times the average "life" of a carbon atom in the biosphere, if a steady state exists. Estimation of this life is somewhat easier than might appear on first glance since 90 percent of the fixation is by ocean plankton, which are minute organisms of very short life. Furthermore, even the carbon contained in longer-lived organisms does not in general have a "life" equal to that of the organism, but rather the material of the organism is replaced a number of times during its life. A maximum average "life" of a carbon atom of not more than a few years is indicated by these considerations. Since biocarbon is such a small fraction of the total reservoir, a more exact treatment appears unnecessary. Taking 10 years for the average carbon life, our calculated biosphere holdup

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[25] Sverdrup, Johnson, and Fleming, *The Oceans* (Prentice-Hall, Inc., 1946), p. 208.

[26] Moberg, Greenberg, Revelle, and Allen, Bull. Scripps Inst. of Oceanography, Univ. of Cal. Tech. Series 3, 231 (1934).

[27] K. Buch, Acta Acad. Aboensis, Math. et Phys. 11, no. 5 (1938).

[28] Reference 25, p. 250.

[29] Reference 25, p. 191.

[30] Reference 25, p. 250.

[31] K. Buch, Acta Acad. Abonensis, Math. et Phys. 11, no. 12 (1939).

[32] F. Paneth, quoted by G. Kuiper, *The Atmospheres of the Earth and Planets* (University of Chicago Press, Chicago, 1949), p. 1.

[33] W. J. Vernadsky, *Geochemie in Ausgewahlten Kapiteln* (Akademische Verlagsgesellschaft, Leipzig, 1930).

[34] E. Rabinowitch, *Photosynthesis and Related Processes* (Interscience Publishers, Inc., New York, 1945), Chapter I.

[35] G. Schroeder, Naturwiss. 7, 8 (1919); 7, 976 (1919).

[36] J. Riley, Bull. Bingham Oceanographic Coll. 1, 1 (1941), quoted by Rabinowitch, reference 34, p. 6.

[37] Reference 34, p. 6.

will be taken as 0.33 g/cm^{-2}, which will prove to be only 4 percent of the total.

The only way of appreciably increasing the hold-up of the biosphere is by the assumption that a considerable portion of this material spends many years in slow decay as humus or ocean sediments before it is recycled. The available evidence seems to be that the major part of the dead material is dissolved during the settling process and that little of it ever reaches the ocean floor.[38] The possibility of appreciable hold-up in humus can be eliminated on the basis of the comparatively small amount of carbon involved in the terrestrial cycle.

In addition to the dilution of cosmic radiocarbon by the carbon of the exchange reservoir, some is being removed constantly by incorporation in sedimentary rocks as they form. This rate of loss of radiocarbon will be estimated. The rates of deposition of calcium carbonate in the sea are not at all well established. Various estimates can be made on the basis of the total amount of calcium carbonate that has been deposited, the bicarbonate contents of rivers emptying into the sea, and the direct observation of the rate of formation of ocean sediments. For example, Brown[39] has calculated the following inventory of carbon in the sedimentary rocks: in shale, 5.45×10^{21} grams; in sandstone, 0.20×10^{21} grams; and in limestone, 3.67×10^{21} grams; for a total of 9.3×10^{21} grams of carbon. If we assume uniform deposition over a two-billion-year period, we have a yearly loss rate of 5×10^{12} grams, which would remove only 6 grams of the 9800 grams of radiocarbon made each year. This may be low because recycling may be important, and the other methods give somewhat higher figures; but it seems likely that the total removal by the formation of sedimentary deposits is not likely to exceed about 3 percent.

We may now consider the question as to whether or not the various portions of the exchange reservoir are sufficiently well mixed so that they are 100 percent efficient in the dilution process. The uniform distribution of C^{14} throughout the reservoir will result only if the various mixing processes are complete in a time short compared with the average life of C^{14}. From the estimated photo-synthetic fixation rate given above and the total material in the exchange reservoir, it can be calculated that 250 years would suffice to turn over all the carbon through the biosphere. This insures that the thin layer of the reservoir at the surface of the earth and of the ocean will be well mixed. The main question is whether or not the depths of the oceans and the upper reaches of the atmosphere are mixed.

Considering the latter first, we note that Yuan's data indicated that the maximum in the neutron flux intensity occurs at about 40,000 feet and that it has fallen off considerably by 70,000 feet. At this altitude the air pressure is 3.5 mm Hg. Excellent and rapid mixing is well established in the troposphere, the adiabatic portion of the atmosphere in which most weather phenomena occur. In the meteorological "standard atmosphere" the tropopause, or boundary between the adiabatic troposphere and the isothermal stratosphere, is assumed to be[40] at 10.8 km (36,000 feet). However, the height of the tropopause varies considerably with the season and with latitude, reaching 18 km (58,000 feet) in the winter at 40° N latitude while in summer it may be found at 16 km (52,000 feet) as far north as 60° N geomagnetic latitude.[41] Thus the tropopause is actually above the altitude of maximum neutron intensity some of the time each year. Even the isothermal stratosphere is not without vertical mixing.[42]

The situation with regard to oceanic mixing is much less obvious. Very little indeed is known concerning the quantitative rates of convective mixing of the deep ocean basins. (Diffusive mixing is of course so extremely slow as to be out of the question.)

From the limited data available on ocean circulation at moderate depths (*ca.* 4000 meters), one can estimate that mixing in the Atlantic Ocean, for example, may occur on a time scale of the order of 10^3 years, but this estimate is a crude one and not applicable to deep basins.

Another argument in favor of complete mixing of the ocean basins may be based on the following considerations. It is known that heat is being liberated from the earth's crust at the rate[43] of about 30 cal/cm^2-yr. Presumably the evolution of heat from the bottom of the ocean is of a similar magnitude,[44,45] and one might expect regions of temperature inversion to develop near the ocean bottom for this reason, especially in areas of poor circulation. To transfer this amount of heat by molecular conduction would require a thermal gradient of 8×10^{-4} deg/cm (if the conductivity of water is assumed to be 0.0013). If a layer of 1,000-meters thickness at the bottom were cooled by molecular conduction only, a temperature difference of 80°C would be required. Naturally, turbulent eddy circulation patterns would be set up at much lower gradients, and the more effective mechanism of eddy conductivity would operate to remove the heat. Such eddy circulation would be equally efficacious in the transfer of dissolved carbonate, and a very efficient method of maintaining the C^{14} equilibrium would arise. It can be calculated, for example, that a temperature gradient of 1.6×10^{-6} deg/cm would require a coefficient of eddy conduction some 500 times the molecular coefficient for the transfer of the postulated amount of heat. (This is quite small as eddy coefficients go, since they often range up to 10^8 or more times the molecular coefficients.) Such an

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[38] Reference 25, p. 1012.

[39] H. S. Brown, private communication.

[40] H. R. Byers, *General Meteorology* (McGraw-Hill Book Company, New York, 1944), p. 171.

[41] C. G. Rossby in *The Atmospheres of the Earth and Planets*, Ed. G. Kuiper (University of Chicago Press, Chicago, 1949), p. 21.

[42] L. Spitzer, Jr., reference 41, p. 221.

[43] E. C. Bullard, Nature **156**, 35 (1945).

[44] H. Pettersson, Nature **164**, 468 (1949).

[45] A. E. Benfield, Nature **165**, 445 (1950).

eddy circulation operating at depths below 4,000 meters can be shown[46] to be adequate to mix the ocean depths sufficiently so that the assay of C^{14} at 6000 meters would be 90 percent of normal with an integrated effect of only 1.4 percent on the mean specific activity.

Few cases of temperature inversion in the deep ocean have been established. The inversion gradient chosen in the above calculation is that observed in the Mindanao Trench[47] and is almost exactly equal to the gradient which would be produced by adiabatic compression (i.e., there is no density inversion, the density of the water being constant with depth, as is also the potential temperature). However, the smaller the thermal gradient the larger the coefficient of eddy conduction required to transport the heat. Without specifying the nature or source of the circulation, we merely point out that if 30 cal/cm^{-2}-yr are being evolved from the ocean floor, a certain minimum rate of circulation must exist in order to prevent the development of large thermal gradients. The above arguments are of necessity qualitative, since turbulent eddy circulation is not amenable to calculation. However, if the assumption of the specified heat evolution is correct, the absence of appreciable thermal inversions near the ocean bottom would seem to indicate that the mixing is very good indeed on our time scale.

It has not been possible to obtain samples of dissolved carbon from the very deep ocean, so that experimental evidence on the mixing time is not at hand. It would appear that measurements of this kind might be of considerable oceanographic interest and might shed some light on the problem of circulation in the deep basins.

We arrive finally, therefore, at 8.3 g/cm^2 for the value of the diluting inventory, distributed as indicated in Table I through the exchange reservoir. The uncertainty in this value we place at about 15 percent, most of this error being in the estimated uncertainty in the value for ocean carbonate. Combining with the value of 156 ± 30 for the neutron intensity/cm^2-min. we calculate an expected specific activity of 18.8 ± 5 per minute per gram of carbon.

IV. EXPERIMENTAL TECHNIQUE

The measurement technique employed the screen wall counter[48,49] with a sample consisting of elementary carbon and the shielding accomplished by a combination of several inches of lead and iron and a surrounding layer of anticoincidence counters.[50] The sample was disposed on a movable cylinder so that the background could be measured by sliding it to one end of the counters away from the sensitive volume.

The preparation of the sample consisted in direct combustion (or in the addition of acid in the case of carbonates) to produce carbon dioxide, which was then purified by precipitation as calcium carbonate, reaction of this carbonate with acid to form carbon dioxide a second time, and reduction of the purified carbon dioxide with hot magnesium metal.

The samples were washed frequently with dilute acid prior to combustion in order to remove any occluded calcium carbonate which might be decomposed during the combustion. These cleansing precautions were found to be necessary in certain instances and were instituted as a standard step in the procedure in nearly all cases.

The elementary carbon sample (8.5 g) was moistened with 20 cc of 0.15 percent agar-agar solution plus about 30 cc of distilled water to make a paste, which was spread smoothly over the inside of the sample cylinder, dried in a warm air stream until the paste was no longer fluid, placed in the counter which was then evacuated for several hours during which the remaining water was pumped off, and then filled with 0.6 cm of ethylene and 10 cm of argon.

A standard run consisted of 48 hours with alternations of the sample cylinder every 4 to 8 hours between the "sample" and the "background" positions. The relative time spent on the sample and background depended on the difference between them, and the intervals were taken as the square roots of the ratios of the absolute counts under the two different conditions.

The observed difference between the sample and the background together with the Poisson statistical error for both were used to calculate the activity of the sample, together with the error in it. The absolute activity of the sample was determined by a separate calibration of the counter.

For the calibration of the counter, carbon dioxide which had been assayed absolutely in a gas counter[8] was used to prepare a carbon sample, which then was counted in the usual manner; and the percent response was used to determine the absolute efficiency. The result was 5.46 ± 0.03 percent for 8 grams of carbon spread over 400 cm^2.

V. EXPERIMENTAL RESULTS. THE DISTRIBUTION AND MEAN SPECIFIC ACTIVITY

It was planned to select samples from widely scattered points over the earth's surface so that any latitudinal variation caused by the known variation in the neutron component of the cosmic radiation would be revealed. The material chosen for the majority of the samples was wood. This choice was made because wood was thought to be the material most likely to be encountered in the program of archaeological age measurements to which the present research was fundamental. A second point was that wood is widely distributed through great extremes of latitude and altitude, and is readily available from museum collections. The final point was that

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[46] The authors are indebted to Dr. Henry Kierstead of the Institute for Nuclear Studies for his interest in this question and for his assistance in the calculations.

[47] Reference 25, p. 739.

[48] W. F. Libby, Phys. Rev. **46**, 196 (1934).

[49] W. F. Libby and D. D. Lee, Phys. Rev. **55**, 245 (1939).

[50] Detailed blueprints may be obtained from the authors.

TABLE II. Activity of terrestrial biosphere samples.

Source	Geomagnetic latitude	Absolute specific activity (dpm/g)
White spruce, Yukon (Frederick Johnson)	60° N	14.84±0.30
Norwegian spruce, Sweden (Donald Collier, CNH Museum)	55° N	15.37±0.54
Elm wood, Chicago (Authors)	53° N	14.72±0.54
Fraximus excelsior, Switzerland (Donald Collier)	49° N	15.16±0.30
Honeysuckle leaves, Oak Ridge, Tenn. (C. H. Perry, Clinton Laboratory)	47° N	14.60±0.30
Pine twigs and needles (12,000 ft alt.), Mount Wheeler, New Mexico (Robert Fryxell)	44° N	15.82±0.47
North African briar (John Hudson Moore, Inc.)	40° N	14.47±0.44
Oak, Sherafut, Palestine (Donald Collier)	34° N	15.19±0.40
Unidentified wood, Teheran, Iran (M. Hessaby)	28° N	15.57±0.31
Fraximus Mandshurica, Japan (Donald Collier)	26° N	14.84±0.30
Unidentified wood, Panama (John Simpson)	20° N	15.94±0.51
Chlorophora excelsa, Liberia (Donald Collier)	11° N	15.08±0.34
Sterculia excelsa, Copacabana, Bolivia (9000 ft alt.) (Donald Collier)	1° N	15.47±0.50
Ironwood, Majoro, Marshall Islands (Donald Collier)	0°	14.53±0.60
Unidentified wood, Ceylon (Donald Collier)	2° S	15.29±0.67
Beech wood, Tierra del Fuego (Junius Bird)	45° S	15.37±0.49
Eucalyptus, New South Wales, Australia (Donald Collier)	45° S	16.31±0.43
Seal oil from seal meat from Antarctic (Byrd Expedition *via* H. J. Deason)	65° S	15.69±0.30
Average		15.3 ±0.1[a]

[a] Error of calibration of counter raises error on absolute assay to 0.5.

it seems from a chemical point of view that cellulose and lignin should be relatively free from any danger of exchanging their carbon with other materials after the death of the tree, and samples from museum collections could be used even though they had been collected several years ago. Carbonate samples were regarded with suspicion because it was feared that exchange between crystalline calcium carbonate and carbon dioxide might occur in moist air. Only a few measurements of contemporaneous ocean carbonate were made to confirm the expected fractionation of the isotopes.

One group of samples was concentrated near the geomagnetic equator where the neutron flux is a minimum, and another in high latitudes where the neutron flux is a maximum. Some consideration was given to the archaeological importance of the region with the thought that, if no uniformity were demonstrated, these data might be utilized for the age measurement research. Two samples from high altitudes were measured, but the height of the timber line was of course very small indeed compared with the altitudes at which radiocarbon production occurs.

Experimental results are given in Tables II and III, which record the type of material measured, its geographical source, and the observed specific activity in absolute disintegrations per minute per gram of carbon. The associated error is the standard deviation calculated from the counting statistics only.

The average specific activity of all of the biosphere samples is found[51] to be 15.3±0.5 absolute dpm/g. The data seem to show no scatter other than the expected statistical fluctuations, indicating that the samples involved did indeed have specific activities identical within the error of measurement, and that the variation over the earth's surface is within the experimental error of the radioactivity measurement of about 2 percent, also. The data presented in Table III for shell reveal that shell is definitely more radioactive than organic material. This difference is to be expected, since it has been shown[52,53] that fractionation of the stable isotopes of carbon, C^{12} and C^{13}, occurs in these systems. The ratio of the C^{14} content of carbonate to that of organic material which is found here is 1.09±0.03. On the basis of the fractionation factor of 1.03 found for C^{13} against C^{12}, one would expect a value of 1.06 for C^{14}. The difference between this value and the one found in the present investigation is not outside of the present experimental error, and since the accuracy of the estimation of the fractionation factor is better than the precision of our measurements of the shell activity, we take the observed value to be 16.2±0.5, derived by multiplication of the mean of the organic specific activity by 1.06. It seems that a further investigation of the fractionation factor would be worth while.

TABLE III. Activity of contemporaneous shell samples.

Source	Assay
Murex shell, Florida West Coast	17.1±0.5
Freshly deposited ocean sediments, 1150-meters depth, Bermuda (J. Lawrence Kulp)	17.4±0.6
Oyster shells, Chesapeake Bay	15.1±0.5[a]
Average	16.5±0.5

[a] This sample had a high ash (22 percent), so the number is less reliable than the counting error indicated.

In order to calculate the mean world-wide specific activity of carbon in the exchange reservoir, we take the weighted mean using 1.0 g/cm^2 as the total amount of organic material in the reservoir and 7.3 g/cm^2 for the carbonate carbon. The result is 16.1±0.5. The agreement of this value with the expected value of 18.8±5 is very gratifying and suggests that no major factors contributing to the situation have been overlooked, although it does seem likely that the agreement may be in some part due to cancellation of errors.

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[51] Earlier publications have given 12.5 for this number. The counters have been recalibrated for absolute efficiency since, and the new figure is derived from the old one by correction for the more accurately determined efficiency.

[52] A. O. Nier and E. A. Gulbranson, J. Am. Chem. Soc. **61**, 697 (1939).

[53] B. F. Murphy and A. O. Nier, Phys. Rev. **59**, 771 (1941).